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H. B. Radousky
R. L. Reichlin
R. H. Howell

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HIGH PRESSURE DOPPLER BROADENING MEASUREMENTS PERFORMED ON IRON IN A DIAMOND ANVIL*

H.B. Radousky, R.L. Reichlin and R.H. Howell

Lawrence Livermore National Laboratory, Livermore, California

The Doppler broadening spectrum of Fe has been measured to a pressure of 40.0 GPa using a high pressure diamond anvil cell (DAC). This represents the first positron annihilation experiment which utilizes DAC technology. As a function of increasing pressure, the electron momenta of Fe shift to higher values, and the bcc to hcp phase transition at 14.0 GPa is clearly observed.

INTRODUCTION

One goal of high pressure physics is to understand the electronic structure of materials, and how it affects observable physical quantities. To this end, many band structure calculations have been carried out on a variety of different materials at high densities. It has proven very difficult to check these calculated band structures directly, since most of the experimental techniques which are normally used do not work at very high pressures. De-Haas Van Alphen experiments have been performed up to approximately 2.5 GPa (1). Photoemission studies have not been performed at high pressures because of the difficult requirement of a very clean surface.

Doppler broadening and angular correlation techniques can provide detailed information concerning electron momentum distribution in a material. Some early positron measurements have been made on a few simple metals below 10.0 GPa using Bridgeman anvil devices.(2-4) The more recent advent of the diamond anvil cell (DAC) has made experimental studies up to pressures of many tens of gigapascals possible (5) opening up an entirely new range of high pressure phenomena for study.

We have measured Doppler broadening spectra for Fe to 40.0 GPa. Fe was chosen because of the magnetic to non-magnetic structural phase transition (bcc to hcp) near 14.0 GPa (5). Also a second phase change (bcc to fcc) had been observed with positrons in thermal studies (6).

Our results show that as a function of increasing pressure, the electron momenta of Fe shift to higher values. The phase transition fron the bcc to hcp structure is also clearly seen in the Doppler broadening data for iron.

EXPERIMENT

To obtain high pressure we used a Mao and Bell (7) type cell with diamonds which had cutlets of 0.8mm. The sample was a sandwich of ^{22}Na in two annealed, 50 micron Fe foils (99.998 pure). The foils in this geometry are thick enough to stop 85% of the positrons (8). The sample was placed in a 350 micron diameter hole in a hardened type 301 stainless-steel gasket without any pressure medium. A schematic of the diamond cell and sample geometry is shown in figure 1 Small ruby chips (5-20 microns) were placed in the sample chamber for pressure calibration using the ruby fluorescence technique (9). Pressures were measured to better that 0.5 GPa accuracy and pressure gradients were less than 3.0 GPa at 40.0 GPa.

An intrensic Ge detector with about 9% effeciency relative to a 3 x 3 inch NaI was used to measure the Doppler broadening spectrum with a resolution of 1.1 KeV. Our sample preparation techniques resulted in count rates of approximately 50 counts/sec in the 511 KeV peak, allowing us to measure a full spectrum with good statistics (500,000 counts) in less than three hours.

This experiment differs from previous positron work(2-4) at high pressure in that the samples are necessarily small in order to reach 40.0 GPa (.35mm as opposed to 3-8mm in previous experiments) and the source was deposited directly on the sample foils, rather than on a supporting material. The Doppler broadening data were reduced to the ratio of low momentum events to total events, S. Plots of the S parameter as a function of pressure and volume are shown in Figs. 2 and 3. The error bars in the figures were estimated by taking repeated data at various pressures and using the standard deviation as the error.

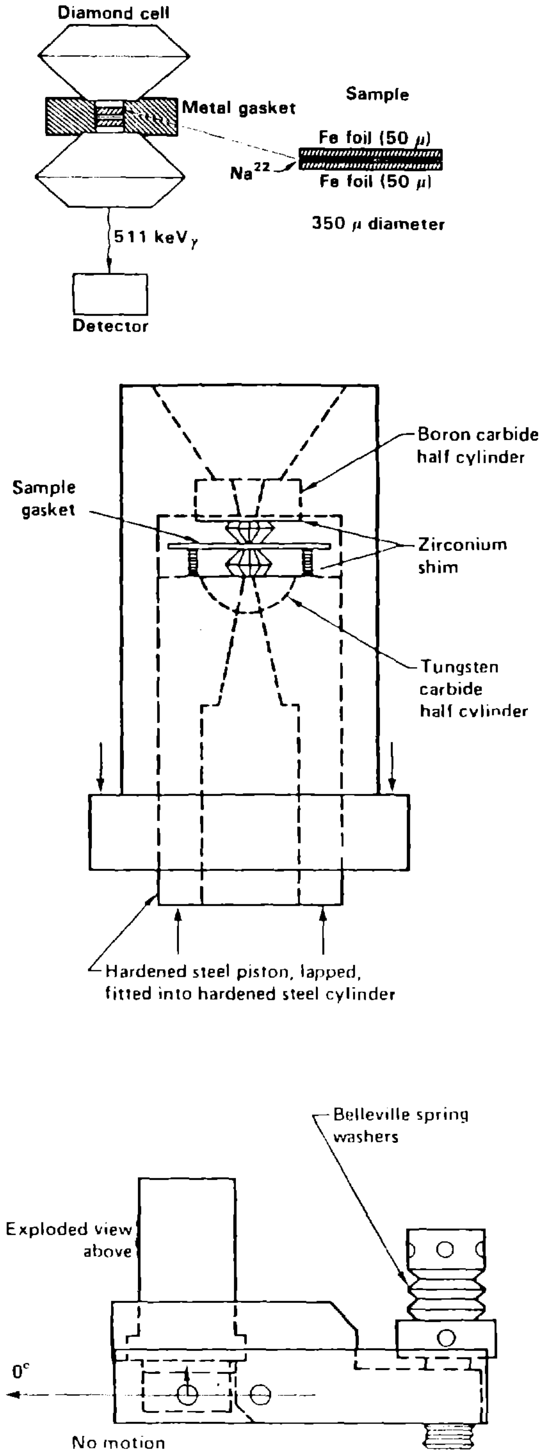

Fig. 1 Experimental configuration from bottom to top the vice, the holder for the diamonds and the details of our samples are shown.

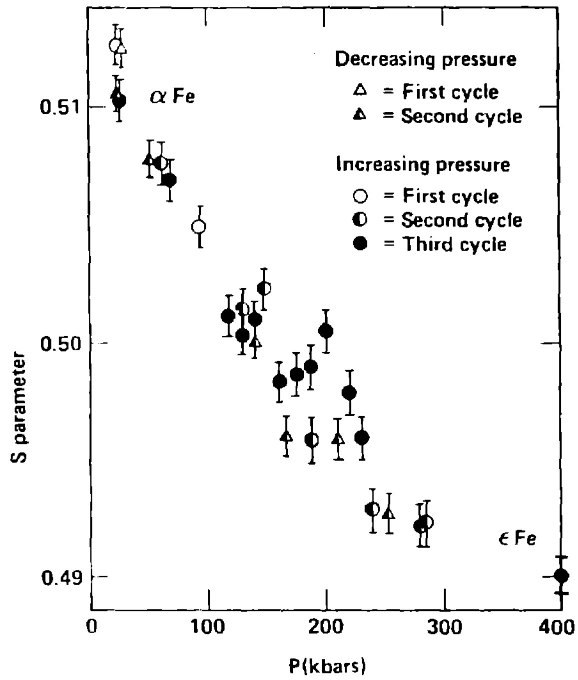

Fig. 2 S parameter vs. pressure. Note the change in slope for bcc and hcp Fe. Shaded area indicates transition region.

Fe is an interesting material to study for two reasons: it is not a simple metal, with d bands playing a crucial role in determining its properties, and it has a structural phase transition near 14.0 GPa from a bcc to hcp structure. The transition has recently been shown to be driven by the magnetic contribution to the free energy (10). The high pressure hcp phase is non-magnetic at room temperature.

The plot of the S parameter vs. pressure shown in Fig. 2 can be visually divided into three regions. Below the transition pressure, S falls linearly with pressure from 2.4 GPa to the onset of the transition at 14.0 GPa. The transition is sluggish and takes approximately 10 GPa to complete. This has been seen from previous x-ray (11) and resistivity (12) data, and is apparent in our positron data as well. From 14 to 24 GPa the Fe appears to be in a two phase region. This region is characterized by a cusp-like feature in the data which is most likely associated with defects, rather than any intrinsic property of the material. It is interesting to note that a similar cusp-like feature is also observed in the positron spectrum of Fe as it is heated through its bcc to fcc transition near 1300°K (6). While the effects in the two phase region are not understood in detail, comparison with the temperature data indicates that this is a real effect rather than spurious noise in the results. Above 24.0 GPa, Fe appears to be completely in the hcp phase and the data once more show a smooth decrease of the S parameter with increasing pressure.

The decrease of S with pressure indicates that the electron distribution is shifting to higher energies at high pressures. As volume decreases, the Fermi energy moves to higher values, causing the positron annihilation rate with valence electrons of higher energy to increase. In addition, the fraction of core annhilation increases as the volume is compressed. These results are consistent with those found previously at lower pressure for Na (3,4) and Al (2).

One interesting feature of the data is that the rate of change of S with pressure in the bcc and hcp phases is markedly different, indicating that our data are sensitive to changes in the electron momentum distribution due to other effects such as the loss of magnetism (10,13) or changes in the Fermi surface topology (14).

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